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An Electromagnetic Squeezer for Compressing Squeezable Electron Tunneling Junctions

by

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The resistance of squeezable electron tunnel junctions (SET junctions) can be adjusted with an electromagnetic squeezer. For junctions immersed in liquid helium, the resistance is stable to approximately 0.1%. This stability is sufficient for measurements of superconducting energy gaps and for superconducting phonon spectroscopy out to 50 mV applied bias. Increased stability, especially at higher biases, will be necessary for inelastic electron tunneling spectroscopy

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Abstract

The resistance of squeezable electron tunnel junctions (SET junctions) can be adjusted with an electromagnetic squeezer. For junctions immersed in liquid helium, the resistance is stable to approximately 0.1%. This stability is sufficient for measurments of superconducting energy gaps and for superconducting phonon spectroscopy out to 50 mV applied bias. Increased stability, especially at higher biases, will be necessary for inelastic electron tunneling spectroscopy.

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Introduction

Electron tunneling junctions have been extensively investigated theoretically and experimentally. 1-5 The majority of these junctions rely on the formation of a thin, native-oxide tunneling barrier between two metal electrodes. Giaever's original experiments on these junctions formed the foundation for many later works that include mechanisms and applications of superconductivity, 1,7 molecular spectroscopy of surface absorbates 4,8 and visible light emission. Electron tunneling has also been studied in Schottky barrier contacts 10 and Esaki tunnel diodes 11 where it has proven to be a sensitive probe of the semiconductor's phonon spectrum.

Since these systems depend on the formation of an oxide barrier or the proper doping of a semiconductor, their applications are limited. The applications would be extended if an arbitrary insulating material could be used as a tunneling barrier. Great progress has been made by a series of researchers in "vacuum tunneling" between a point or a small sphere and a surface. Binnig et al. 12 are capable of resolving surface topgraphy to within 0.1 Å. Spectroscopy, however, requires setting a tunneling barrier gap at several angstroms stable to less than 0.01 Å. We utilize a method that employs a squeezable electron tunneling (SET) junction that has not only been used to observe superconducting energy gaps 13 but is stable enough to obtain phonon spectra of evaporated superconducting Pb and Al films.

These junctions consist of two crossed electrodes evaporated on substrates that are separated by thin film spacers. An electromagnetic "squeezer" is used to compress the gap by flexing the substrates so that tunneling can occur between electrodes. Concaivably, this squeezing force could be generated many ways but the electromagnet adapts easily to low temperature experiments and can apply a large range of forces. The total force necessary to compress the junction and its vibrational stability depend on the junction spacer thickness and geometry, the substrate thickness and modulus of elasticity, the materials in the gap, electromagnet current fluctuations and squeezer construction.

A test for the existence of a tunneling current is the presence of a superconducting energy gap at low junction bias. Once observed, additional verification can be inferred from structure in the I-V characteristics due to the interaction of tunneling electrons with the normal modes of the junction. These normal modes include metal and barrier phonons and molecular vibrations of impurities trapped in the barrier. Since the structure in the I-V characteristics due to these modes is typically small, it can only be seen if the junction is stable enough. As a rough guide: seeing a gap requires resistance stability better than 10%; superconducting phonons, better than 1%; molecular vibrations, better than 0.1%.

We report here the beginning of such a study using SET junctions compressed with an electromagnetic squeezer. Superconducting energy gaps and phonon spectra are clearly visible for Pb-Pb and Al-Pb junctions immersed in liquid helium. These results show the squeezer's capabilities and limitations

and provide insight for improvement that may allow molecular vibrations to be seen.

Apparatus

Dust particles caused microshorts or prevented tunneling from taking place, ruining several initial experiments. Therefore, care was taken to avoid dust by cleaning subtrates (1" x 3" x 1 mm glass microscope slides) and assembling the junctions in a laminar flow hood that enclosed the opening of the vacuum system in which the electrodes were evaporated. The slides were first hand washed with a solution of Liquinox and rinsed with tap and then deionized water. They were then transferred to the laminar flow hood and stored under water until needed. Just prior to an evaporation, they were rinsed with purified water that had been filtered for 0.2 µm particles and then degreased and dried in acetone vapor.

Two electrodes (\approx 0.1 μm thick) and four spacers (\approx 0.9 μm thick) were evaporated onto the slide in the diffusion pumped system at a pressure of 10^{-3} Pa and at a rate of 10 Å/S. The slide was removed from the vacuum system, cut into two 1" \times 1.5" pieces and assembled by hand into a SET junction as shown in Fig. 1. Transparent plastic tape holds the slides together and prevents dust from entering the junction area.

Two precautions were taken to insure that the average gap was nearly constant over the area of intersection. First, this area was small in that the electrodes narrowed to 50 um at the crossing point. Second, the substrates were flexed by applying opposing forces along concentric circles outside and inside

of the spacers on the two sides of the junction. This way, the substrates moved in the same direction but would bend with slightly different radii of curvature.

To quantify the spacing as a function of position in the intersecting area, the junction may be approximated as two thin concentric discs having a thickness, t, modulus of elasticity, E, and reciprocal Poisson ratio, m, separated by a spacer ring having a radius, a, and thickness, h. If a force, F, is applied along circles of radii r_i and r_0 illustrated in Fig. 2, then the separation as a function of r, the distance to the center of the junction, is,

$$Z(r) = Z(0) + \delta Z(r)$$

where

$$2(0) = h - \frac{3F(m^2-1)}{2\pi Et^3 m^2} \left[a^2 \ln \frac{r_0}{a} - r_1^2 \ln \frac{a}{r_1} + \frac{3m+1}{2(m+1)} (2a^2 - r_0^2 - r_1^2)\right]$$

and

$$\delta Z(r) = r^2 \times \frac{3r(m^2-1)}{2\pi E t^3 m^2} \left[\ln \frac{a^2}{r_1 r_0} + \frac{m-1}{2(m+1)} \left(\frac{a^2}{r_0^2} - \frac{r_1^2}{a^2} \right) \right].$$

For a SET junction composed of 1 mm thick glass substrates of radii r_1 = 0.2 cm and r_0 = 1.4 cm and a spacer radius of 1.0 cm, a force of 1 N is necessary to decrease the gap 0.7 um so that tunneling can occur between 0.1 um thick electrodes. This is assuming m = 4 and E = 7 x 10^{10} N/m², the handbook value for glass. 15

Fig. 2 also shows a plot of $\delta Z(r)$ under this condition. If the electrode intersection area extends 30 um from the center of the junction, then the variation is less than 0.1 Å! Moreover, if the intersection area is not at the center but a

variation of 1.0 Å can be tolerated, then the electrodes should cross within 150 μm of the closest approach of the substrates. In comparison, similar calculations for a central point loading of the top substrate and a fixed bottom substrate using the same junction parameters above show that the barrier variation is roughly twenty times greater and that the electrodes must intersect within a few μm of r=0 for the total variation to be less than 1 Å. We emphasize the approximate nature of these calculations presented only to illustrate the advantages of this squeezing geometry. In fact, most of the microscope slide substrates were warped more than a μm over their surfaces so that our SET junctions required forces ranging from 1 to 20 N. In some cases, the glass fractured before the electrodes were close enough for tunneling.

Fig. 1 is a photograph of a Pb-Pb SET junction with air in the gap after cycling to low temperatures. The somewhat "moth-eaten" appearance of the electrodes is due to moisture condensation upon warming. Note the interference fringes (Newton's rings) delineating the spacing between the substrates. The inner dark fringe surrounding the electrode crossing point extends a few mm from the center of the pattern. The center dark fringe occurs where the substrates are separated by less than a quarter wavelength which is of order the combined thickness of the electrodes.

It should also be emphasized that the electrodes are not smooth on anything approaching a 1 Å scale. Roughness is probably of order 10s or even 100s of Å. Tunneling occurs between high points on one electrode and high points on the

other. The purpose of having the average spacing nearly the same across the junction is to avoid having all the tunneling (and, hence, all the heat dissipation) at one corner of the junction.

Fig. 3 illustrates the design of the junction squeezer. It consists of two copper coils secured in an iron yoke covered by a moving iron endplate through which the force is applied to the junction. The force is produced by passing a currents through the coils generating a magnetic field in the space between the yoke and the endplate. Since the space is adjustable, different force ranges are possible for a given current range.

For a small space, the force is roughly proportional to

$$F \propto (\frac{Ni}{d})^2$$

where N, i and d are the number of turns in the coil, the current in the coil and the thickness of the space, respectively. For the above mentioned junction geometry, a current of 500 mA and spacing of d = 1 mm depresses a 0.7 µm gap to tunneling distances. Under these conditions, the magnetic field between the yoke and the endplate was measured using a Hall probe and found to be approximately 1000 gauss. The measured field near the junction was less than a gauss, however, due to the low reluctance of the iron surrounding the windings. Purified iron (< 0.05% impurities) was used to minimize hysteretic effects and increase the magnetic permeability.

One of the copper coils has 700 turns and the other has 100 turns both of \$28 gauge wire. Parallel superconducting coils with the same number of turns were later added to

minimize heat dissipation for squeezer operation at lower temperatures (< 1.5° K). The majority of the force is applied by the larger coil with the smaller used as part of a feed-back system that stabilizes the I-V characteristic of the junction. This is useful for junctions with vacuum or gas in the gap.

Since fine force adjustments displace the gap fractions of angstroms, it is important for the plunger mechanism to have minimum friction. This is accomplished by having an oversized clearance hole in the yoke for the plunger assembly. To prevent tilting, the plunger rests on the yoke with the junction held in place by the gap screw so that even if the endplate is not perfectly parallel to the yoke the force will be directed against the junction by pivoting about the edge of the plunger.

In general, junction stability is governed by squeezer current fluctuations along with vibration isolation, viscosity of the fluid in between the substrates and the geometrical parameters of the junction. Of these, we believe the most important to be the latter two since little precaution is taken to avoid vibration. For example, preliminary measurements for Pb-Pb SET junctions at 4.2° K with a solid such as napthalene melted into the barrier have I-V curves stable to within 0.02% whereas those for liquid helium in the barrier are only 0.1% stable. According to Teague¹² for low bias, the resistance of an idealized tunnel junction with a parallel plane geometry

$$R = \frac{s^2 R_0}{A} \exp(asw^{1/2}),$$

where S, W and A are the barrier thickness, electrode work function and junction area, respectively ($R_0 = 160 \text{ K}\Omega$ and a = $10.25 \text{ (eV)}^{1/2} \text{ nm}^{-1}$). Using a 4 eV work function and a junction area of 2500 μm^2 for nominal tunneling distances between 1.0 and 2.0 nm, an order of magnitude change in the resistance occurs for a 0.1 nm change in S. From this, we infer an average barrier stability of 0.001 Å when resistance fluctuations are on the order 0.1% of the total low bias resistance.

Current versus voltage characteristics were determined using a differential tunneling spectrometer. $\frac{16}{dT^2}$ was measured as a function of voltage by applying a 1120 Hz modulating signal across the junction and measuring the second harmonic voltage with a lock-in amplifier. Traces were taken at low temperatures by placing the experiment in a double insulated dewar filled with liquid helium. The junctions were set after cooling so that differential thermal contractions would not harm the electrodes by mashing the substrates together.

Results

Fig. 4 shows the experimental I-V characteristic obtained when a SET junction consisting of Pb electrodes evaporated on glass substrates was compressed using the squeezer. The apparatus was immersed in liquid helium that was pumped to a

vapor pressure of 600 mTorr so that the temperature was about 1.2° K. The inset shows current versus voltage curves obtained for different applied forces. Notice that the energy gap had the appropriate value for a Pb-barrier-Pb tunnel junction of $2\Delta_{\rm Pb}=2.8~{\rm meV}^{1.7}$ and that it was independent of the applied force. The normal tunneling resistance (V \geq 2.8 mV) was changed from 100 k Ω to 1 M Ω by decreasing the coil current 0.5 ma from an initial value of about 500 ma. The current was adjusted by hand while watching the I-V trace on an oscilloscope. We estimated the leakage current from the conductance inside the energy gap and found it to be less than 1% of that outside the gap for the range of forces shown. Junctions with resistances lower than 10 k Ω had leakage currents much larger, particularly for resistances below 1 k Ω where superconducting microshort(s) were apparent near zero bias.

The acoustic phonon structure observed previously in Pb-PbO $_{\rm X}$ -Pb tunnel junctions was also visible in the second harmonic versus voltage curve as seen in Fig. 4. In fact, our results were very similar to those of Rowell and McMillan in that the dips associated with the TA and LA phonons of Pb showed additional structure due to Van Hove singularities in the phonon spectrum. 18

Fig. 5 shows the current versus voltage curve at 1.2° K for a SET junction consisting of Al and Pb electrodes evaporated on glass substrates. At this temperature, both the Pb and the Al films were superconducting so that the energy gap structure should have been located at $\Delta_{\rm Pb} = \Delta_{\rm Al}$. A current source was used to take the I-V trace so that the negative resistance

region near the gap edge was obscured. As the junction warmed, $2\Delta_{\rm Al}$ went to zero around 2° K. Using this as T_C for a 3CS superconductor, $2\Delta_{\rm Al}$ should theoretically be 0.4 meV, very close to our observed value. ¹⁹

Fig. 6 shows $\frac{d^2v}{dI^2}$ for voltages from 15 to 50 mV across the Al-Pb SET junction. Below 15 mV, Pb phonon structure was similar to that found in the Pb-Pb junctions. In the figure, dips corresponding to the TA and LA phonons of superconducting Al can be observed near 27 and 30 mV, respectively. Again, this had previously been noted near these energies in Al-AlO_X-Pb junctions. On Note that there is a marked increase in the harmonic signal noise for voltages greater than 35 mV. This is typical of these junctions and unfortunate. It is unfortunate in that tunneling spectroscopy of organic molecules demands junction stability out to 500 mV. Nevertheless, there is hope that with junction electrode changes, geometry changes, vibration isolation and a new squeezer design that these instabilities may be overcome.

Conclusion

In summary, an electromagnetic SET junction squeezer was successfully operated at 1.2° K.

i) Superconducting energy gaps were observed in the I-V curves for evaporated Pb-Pb and Pb-Al junctions and yielded a value of $t_{\rm Pb} = 1.4$ mV. The estimated leakage current was less than 1% of the normal current. Structure at $t_{\rm Pb} = t_{\rm Al}$ was present in the I-V curve of an Al-Pb SET junction due to the superconducting Al film ($t_{\rm C} \approx 2.0^{\circ}$ K).

- ii) The second harmonic voltage versus applied bias data showed dips caused by Pb and Al phonons previously observed in metal-oxide-metal junctions. The Pb phonon spectra was very similar to that measured by Rowell and McMillan.
- iii) Junctions were constructed on microscope slides using standard vacuum evaporation techniques supplemented by careful substrate cleaning and dust minimization. The electromagnetic squeezer easily adapted to a liquid helium dewar. Also, phonon spectra were taken without feed back circuitry and with minimal vibration isolation making the entire process experimentally straightfoward.

The present squeezer then satisfies requirements necessary for experiments concerning superconductivity and electrode phonon spectroscopy. With this initial effort, we have gained insight about the exciting capabilities of this technique and hope to increase its range of application to higher electron energies. Perhaps a combination of improved vibration isolation, new junction geometries, smoother electrodes and new squeezer designs will be helpful.

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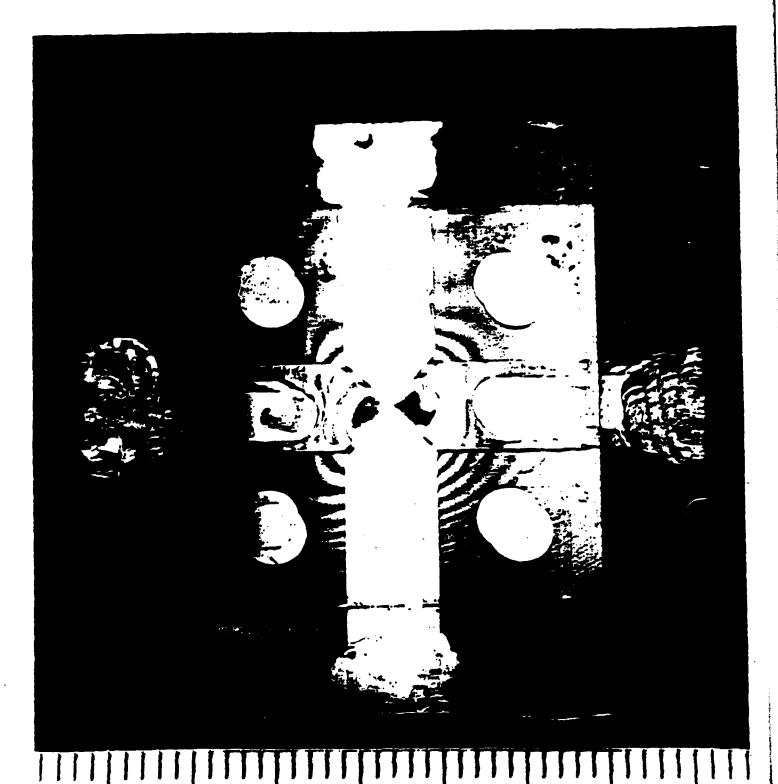
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Figure Captions

- Fig. 1. Photograph of a Pb-Pb SET junction after cycling to low temperatures. Note the four spacers and the junction between narrowed electrodes. Newton's rings delineate the spacing between the substrates (cm scale).
- Fig. 2. Estimated variation of the barrier 6Z(r) near the center of a compressed SET junction. Here, $r_1=0.5$ cm, $r_0=2.8$ cm, a=2 cm, the electrodes are 0.1 μ m thick and the spacer thickness is 0.9 μ m.
- Fig. 3. SET junction squeezer.
- Fig. 4. Second harmonic voltage versus bias voltage for a Pb-Pb SET junction immersed in liquid helium (V_{mod} = 200 μV). The inset shows I-V curves obtained with different forces applied to the junction.
- Fig. 5. The I-V curve of an Al-Pb SET junction immersed in liquid helium.
- Fig. 6. Second harmonic voltage versus bias voltage for an Al-Pb SET junction immersed in liquid helium (V_{mod} = 2 mV).

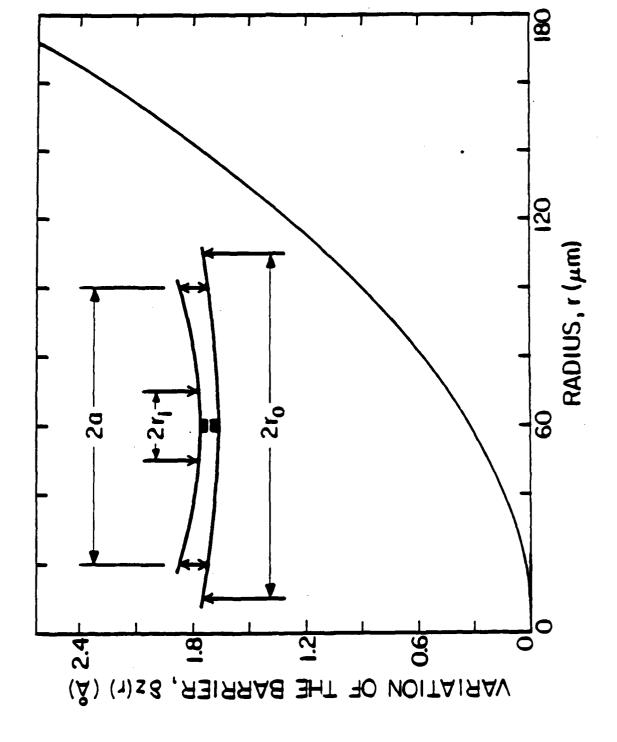


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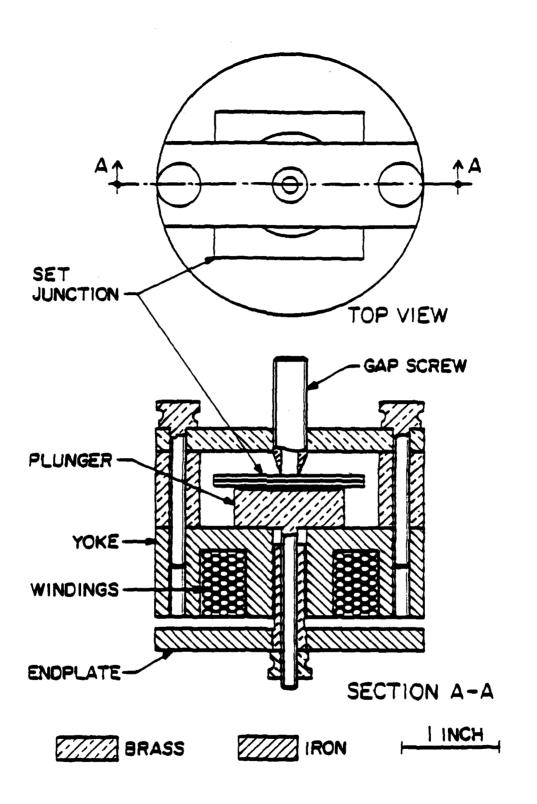
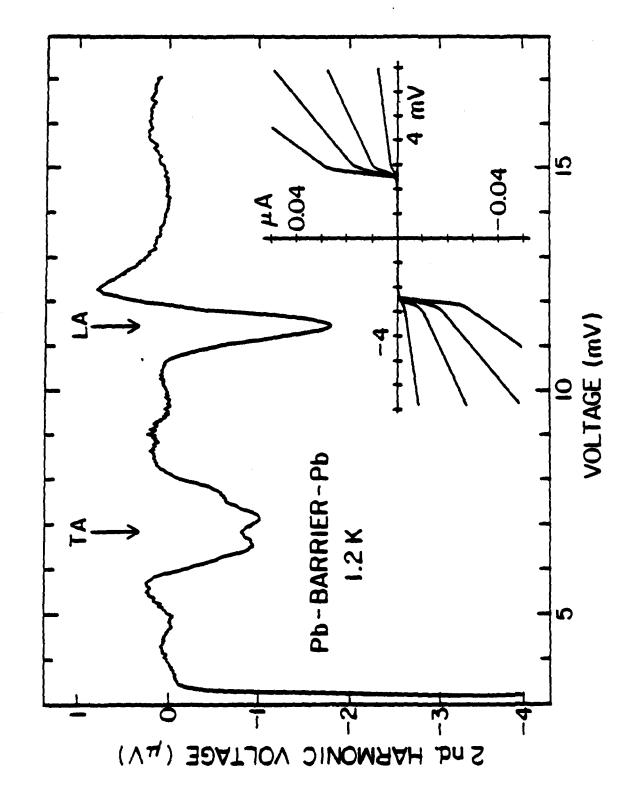


Fig. 3
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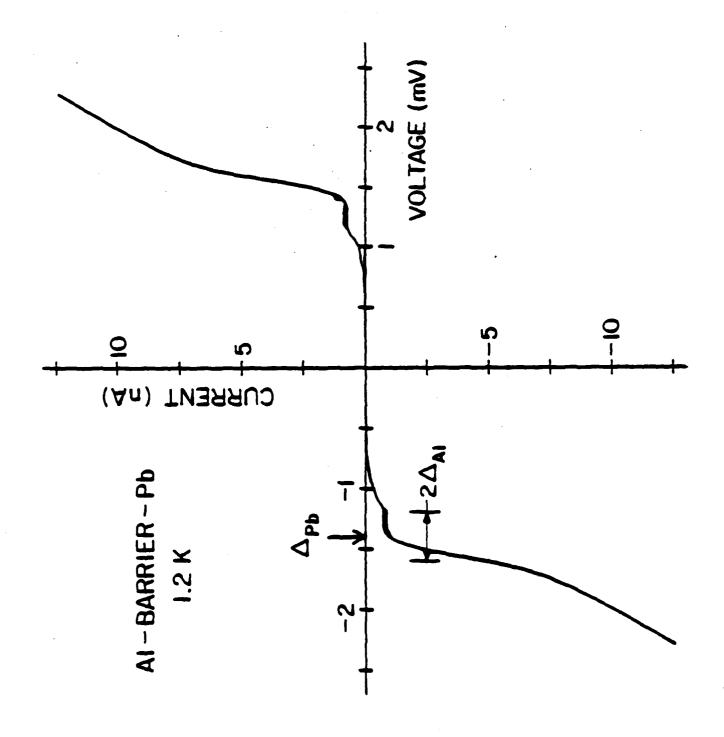


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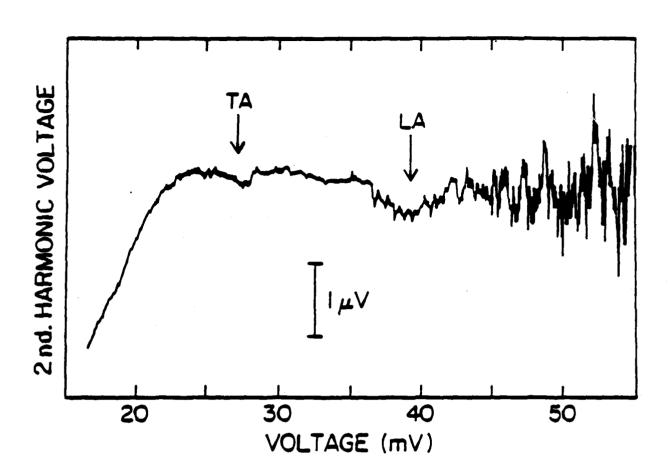


Fig. 6
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